Available online at www.sciencedirect.com

ScienceDirect

Energy Procedia 148 (2018) 599–606

Energy Procedia

www.elsevier.com/locate/procedia

73rd Conference of the Italian Thermal Machines Engineering Association (ATI 2018),
12–14 September 2018, Pisa, Italy

Integration of oxygen transport membranes in glass melting furnaces

Antonio Giuffrida[a]*, Paolo Chiesa[a], Francesca Drago[b], Luca Mastropasqua[a]

[a]*Politecnico di Milano - Dipartimento di Energia, Via Raffaele Lambruschini 4A, Milano 20158, Italy*
[b]*Ricerca sul Sistema Energetico - RSE S.p.A., Via Raffaele Rubattino 54, Milano 20134, Italy*

Abstract

Glass manufacturing is an energy-intensive process where the bulk of energy, necessary to maintain high temperatures for glass melting, usually comes from natural gas combustion. A solution for energy saving is an oxy-fuel glass furnace.
This paper investigates the possible application of a membrane based oxygen separation module in glass melting furnaces. The mass and energy balances of two oxy-fuel glass furnaces have been reproduced and compared to an air-blown unit assumed as benchmark. The two oxy-fuel cases differ for the technology adopted to supply oxygen: in the first case, it is delivered by a PSA/VSA unit, whereas in the second case, oxygen is separated from a hot air stream in a membrane module integrated in a micro-gas turbine system. The assessment of the primary fuel demand of the three options highlights the superiority of the oxy-fuel glass furnace with the advanced membrane-based technology. The economic assessment, included to highlight the potential of the proposed application, outlines further advantages.

Keywords: Glass furnace; Heat balance; Oxygen transport membrane; Energy saving; Economic analysis.

---

* Corresponding author. Tel.: +39-02-23993904; fax: +39-02-23993913.
E-mail address: antonio.giuffrida@polimi.it

10.1016/j.egypro.2018.08.147

1. Introduction

A glass melting furnace can be regarded as a chemical reactor producing glass by a series of solid-liquid state reactions. All glass formation reactions occur at very high temperature (even higher than 1400-1500°C) in a confined space surrounded by refractory. The heat input for melting the batch is supplied by oil or natural gas combustion. Electric booster systems can be installed to supply additional energy to increase the melting capacity. However, only a portion of the total heat input is used for melting the glass. As a matter of fact, most of the heat is carried with the exhaust gas and lost to the walls, the crown and the opening holes of the furnace.

The conventional arrangement of a glass furnace is shown in Fig. 1 [1], where a regenerative heat exchanger consisting of two refractory chambers is present. One chamber is used to absorb and store heat from the flue gas, at the same time the other chamber is used to preheat the combustion air. The glass furnace is installed with a single raw material continuous feeding port called “doghouse” and the regenerator chambers are called as north side (non-doghouse side) and south side (doghouse side).

Energy benchmarking for glass furnaces has been thoroughly discussed by Beerkens [2,3]. Referring to the most recent paper [3], Fig. 1 shows the energy efficiency ranking of 168 existing container glass furnaces, starting from the lowest to the highest specific energy consumption. The energy values are normalized for the case of 50% recycling cullet in the batch, so the total primary energy consumption per ton of molten glass accounts for the primary energy required for electric power generation (in case of electric boosting) and oxygen production. According to the data in Fig. 1, the minimum achievable energy consumption level for normal batch would be around 3.8 MJ/kg, which reduces to around 2.5 MJ/kg for 100% cullet.

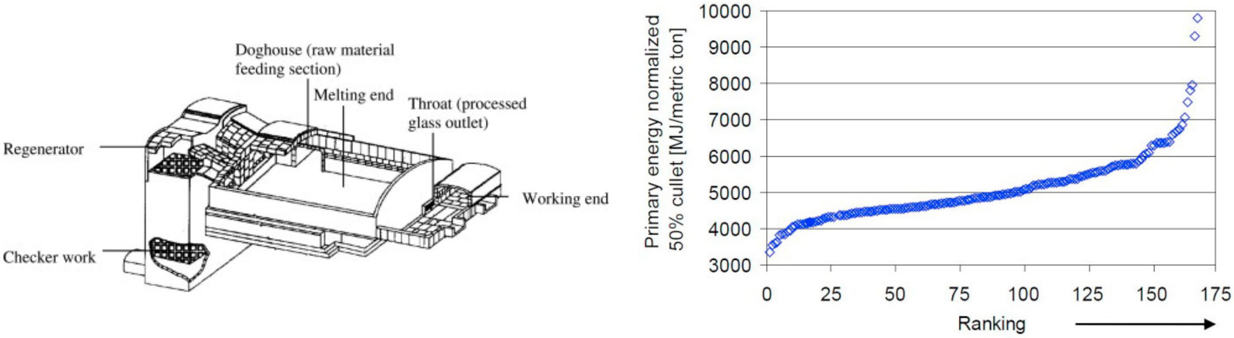


Fig. 1. Glass furnace details, on the left [1], and example of energy efficiency ranking of a set of 168 container glass furnaces, on the right [3].

Oxygen-firing is an effective solution for saving energy in glass furnaces. The Beerkens’ benchmark study [2] reports values of 3.4-3.6 MJ/kg for oxy-fuel furnaces with cullet and/or batch preheating, also including the energy demand for oxygen production.

As a matter of fact, the oxidant in current oxy-fuel glass furnaces comes from conventional technologies, i.e. pressure or vacuum swing adsorption (PSA/VSA) and even cryogenic distillation processes. After reproducing the mass and energy balances of an air-blown glass furnace selected as the case study, this paper proposes an oxy-fuel solution, where the oxygen is produced by a membrane-based system integrated in a micro-gas turbine included in the overall furnace layout. Ultimately, an economic assessment is proposed to highlight the potential of such an application.

2. The case study and the calculation tools

In this paper, reference is made to the work by Sardeshpande et al. [1]. Based on field analyses and experimentations in the glass industry, the authors provide the design and operating parameters of a case study representing the current state of the art of glass furnaces. The heat inputs and outputs for the considered glass furnace are thoroughly analysed and illustrated by means of a Sankey diagram. In detail, the furnace capacity and draw are fixed at 100 and 90 ton/day, respectively, whereas the cullet is set at 40%. Comparing this case study with the ones included by Beerkens in his database [3], it is possible to appreciate that it is a very efficient benchmark, even considering the actual cullet (40% vs. 50%), as the energy input amounts to 3833 kJ/kg.

Starting from the work by Sardeshpande et al. [1], the mass and energy balances have been estimated with GS, a computer code in-house developed in the past years at the Department of Energy of Politecnico di Milano [4].

When addressing the new oxy-fuel furnace solution, a detailed membrane model is employed to propose a reactor design which could fit the process operating conditions derived from GS. It is a 1-D model capable to evaluate the permeation profile of a planar mixed ionic electronic conducting (MIEC) membrane module by integrating the conservation equations along the axial direction by a finite difference method. The model, in-house developed at Politecnico di Milano, has already been employed for the simulation of solid oxide fuel cells and membranes included in small scale catalytic partial oxidizers for hydrogen production from natural gas [5]. A schematic representation of the model adopted for the simulation of supported membranes is depicted in Fig. 2, with four main oxygen transport steps:

- gas diffusion in the feed side, from the gas stream to the membrane wall;
- diffusion across the active ceramic membrane, which comprises: oxygen reduction on the feed side, oxygen-ions bulk diffusion and oxygen recombination on the permeate side;
- oxygen transport in the porous support structure;
- gas diffusion in the permeate side, from the support wall to the gas stream.

The permeation model equations are not reported here for the sake of brevity, but can be found in detail elsewhere [5].

Disc-shaped membranes and supports have been tested at the laboratories of RSE and measured values of oxygen flux through the supported membrane and the supports alone are thus available. The tested membranes are characterised by a $La_{0.6}Sr_{0.4}Co_{0.2}FeO_{3-\delta}$ (LSCF) 30 µm thick active layer, supported on a 0.7 mm porous structure of LSCF. The porosity of the support structure is known and equal to 0.45. The membrane active area is 147.4 mm^2.

In summary, experimental data available at 950°C, 900°C and 850°C and different feed and sweep streams at atmospheric pressure have been used to calibrate the unknown parameters of the OTM model.

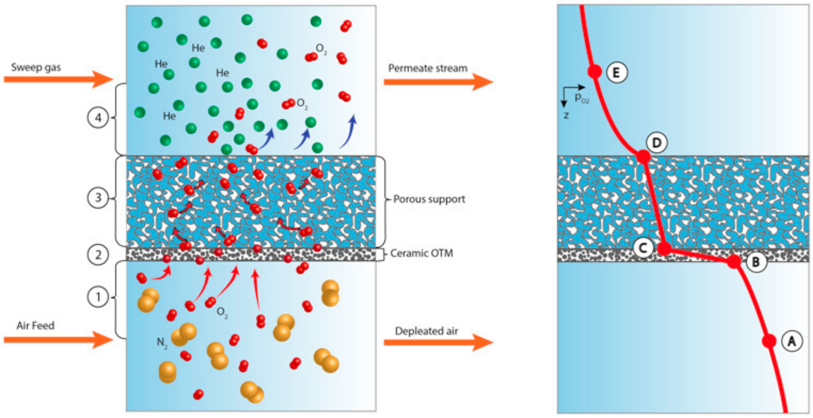


Fig. 2. Schematic representation of the membrane module and the associated O_2 partial pressure profile

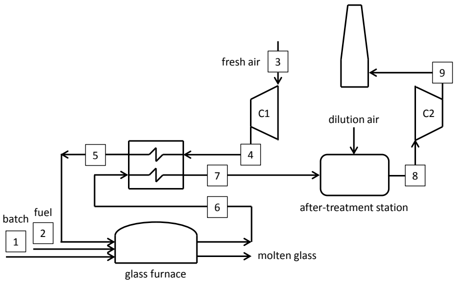
3. Mass and energy balances

This section deals with three cases of glass melting furnace, proposing a simplified layout of the system, the results of mass and energy balances in terms of mass flow rates and temperatures, as well as the results of the related energy consumptions. Natural gas (CH_4: 89%, C_2H_6: 7%, CO_2: 2%, C_3H_8: 1.11%; N_2: 0.89%) with a lower heating value of 46.48 MJ/kg is always considered as the fuel input.

3.1. The reference case

The first case considered is the benchmark, as anticipated in section 2. Starting from the work by Sardeshpande et al. [1], the mass and energy balance of the air-blown furnace has been assessed in order to determine mass flow rates and temperatures. By assuming a combustion air flow rate resulting in 2% for the O_2 content in the gas exiting the furnace [1], Fig. 3 shows a schematic of the system along with the main results of the mass and energy balances.

A significantly larger stream is observed at station 7 compared to station 6 because of air infiltration [1]. In the case of no air infiltration, a temperature of 610.3°C would have been calculated, inclusive of the heat losses from the regenerator. The after-treatment station for the gas exiting the furnace provides for cooling by dilution with ambient air down to 150°C, then the resulting stream is de-dusted and scrubbed before being blown to the chimney.

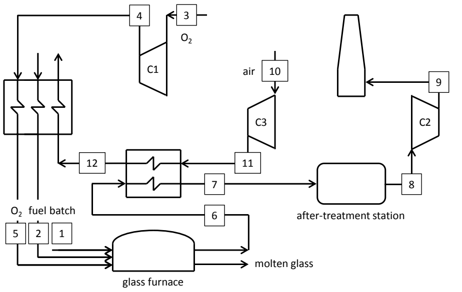


stream	ṁ, kg/h	T, °C
1	3750	15
2	309.3	15
3	5559.8	15
4	5559.8	21.4
5	5559.8	935.2
6	5869.1	1313.7
7	8732.9	361.8
8	24828.1	46.6
9	24828.1	58.1

Fig. 3. Schematic of the air-blown reference furnace and results of mass flow rates and temperatures at the main stations.

3.2. The oxy-fuel case with heat recovery

When switching to the oxy-fuel technology, some considerations are necessary. Beerkens and Muysenberg [6] reported that an efficient oxy-fuel glass melting furnace requires 16.7% less fuel than a similar air-blown case. However, such a saving does not take into account the corresponding energy consumption required for oxygen production. Therefore, with reference to the former calculations of the air-blown reference case, the thermal heat loss of the furnace has been tuned for the same production capacity (90 tons per day) with the lower fuel consumption [6]. In detail, the heat loss has been reduced without varying the temperature of the gas leaving the furnace. On the other hand, some improvements are always possible if considering that an oxy-fuel glass melting furnace is not usually regenerative.



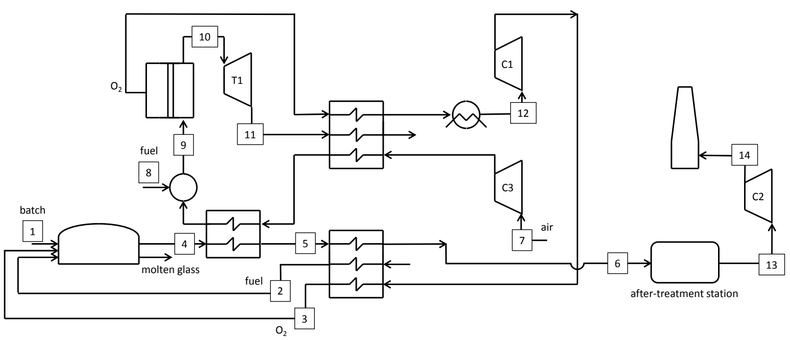
stream	ṁ, kg/h	T, °C
1	3750	15
2	235.7	450
3	983.2	15
4	983.2	21.5
5	983.2	550
6	1219	1313.7
7	1219	835.3
8	10804	50.9
9	10804	62.4
10	1306.4	15
11	1306.4	25.6
12	1306.4	750

Fig. 4. Schematic of the oxy-fuel furnace with heat recovery and results of mass flow rates and temperatures at the main stations.

Inspired by the HotOxyGlass project [7], Fig. 4 shows the schematic of an oxy-fuel glass furnace where air (11) is pre-heated in a recuperator by the high-temperature gas exiting the furnace (6). This hot air stream (12) is then directed to other heat exchangers for pre-heating both the oxygen and the natural gas. In this new case, according to the previous calculation assumptions, a further reduction in natural gas consumption is achieved, with an overall fuel saving of 23.8% compared to the benchmark in Fig. 3. Although such a fuel saving does not include the primary energy related to the oxygen production, this result is absolutely consistent with the saving declared by the HotOxyGlass project [7], i.e. 25% ± 2%. Of course, the final saving will be lower after accounting the necessary energy contribution for oxygen production. Results in Fig. 4 have been calculated based on no air infiltration in the sub-atmospheric pressure zones of the plant, for the sake of simplicity.

3.3. The oxy-fuel case with oxygen transport membrane

The third case investigated in this paper considers a membrane-based system for separating the oxygen from a hot air stream. In detail, the membrane-based system is integrated into the layout of the glass furnace with heat exchangers and turbomachinery, namely a micro-gas turbine system. In this new case, it is possible to deliver the oxygen necessary to the furnace as well as supplying net electric power. The proposed case specifically considers a three-end membrane module with no sweep stream fed to permeate side.



stream	$\dot{m}$, kg/h	T, °C
1	3750	15
2	234.4	450
3	893.6	550
4	1128	1313.7
5	1128	800
6	1128	501
7	8690.4	15
8	57.1	15
9	8747.6	950
10	7853.8	950
11	7853.8	648
12	893.6	40
13	5583.6	58.3
14	5583.6	69.8

Fig. 5. Schematic of the oxy-fuel furnace with oxygen transport membrane integrated into the system and results of mass flow rates and temperatures at the main stations.

The layout for such an advanced glass furnace is schematized in Fig. 5. An air stream (7) is delivered by a compressor through a first recuperative heat exchanger for a preliminary pre-heating (up to 600°C) by the hot gas exhausted from the turbine T1 and by the high-temperature oxygen separated at the membrane system. A second heating up to ~700°C occurs in another heat exchanger by the high-temperature gas (4) exiting the glass furnace. This hot air stream, after the necessary combustion with natural gas to rise the temperature up to 950°C, enters the membrane system at 4 bar with still high oxygen content (greater than 18%). The oxygen separated at the membrane system at an absolute pressure of 0.25 bar (50% of the oxygen present in the feed stream) is partially cooled to pre-heat the compressed air, then further cooled down to 40°C (12). Ultimately, the oxidizer is delivered via a vacuum pump through a recuperative heat exchanger, where it is pre-heated (up to 550°C) together with the natural gas required at the burners, to the furnace. The flow rates in Fig. 5 have been calculated under the hypothesis of no air infiltration in the sub-atmospheric pressure zones of the plant, similarly to the case in Fig. 4.

The details of the main steams in Fig. 5 for the newly proposed glass furnace indicate an overall amount of 291.6 kg/h of fuel, greater than the fuel demand of the case in Fig. 4 due to the fuel required to rise the temperature of the feed stream to the membrane. Nevertheless, the overall fuel demand is reduced by 5.7% compared to the case of the reference furnace in Fig. 3.

3.4. Results and energy consumptions

The results of the energy balances of these three considered cases are reported in Table 1, which details the heat fluxes starting from the fuel energy. The heat carried in the glass is always the same for the fixed production, so the fuel reduction for cases 2 and 3 reflects on a higher percentage compared to the reference case. The heat recovery for case 2 is clearly lower compared to case 1, but both heat losses in flue gas and other heat losses are reduced. According to the previous description, case 3 proposes a solution improving case 2 from an energy point of view. As a matter of fact, heat losses in flue gas are significantly reduced by transferring heat to the compressed air stream before it enters the combustor to finally rise its temperature up to 950°C. On the other hand, the heat recovered and recycled into the furnace is lower compared to case 2, because the oxygen stream delivered by the vacuum pump is warm with a temperature of ~250°C.

Table 1. Details of heat fluxes for the examined cases (1: reference case; 2: oxy-fuel case with heat recovery; 3: oxy-fuel case with oxygen transport membrane).

	1		2		3	
	kW	%	kW	%	kW	%
Fuel energy introduced in the furnace	3992.7	100	3043.5	100	3026.6	100
Heat carried in the glass	1628.1	40.8	1628.1	53.5	1628.1	53.8
Heat recovered and recycled into the furnace	1542.7	38.6	218	7.2	153.6	5.1
Heat loss in the flue gas	900.0	22.5	469.5	15.4	161.6	5.3
Other heat losses	1464.6	36.7	945.9	31.1	945.9	31.3
Heat transferred to the micro-gas turbine cycle	-	-	-	-	290.9	9.6

The energy consumptions of the three considered cases are reported here for a preliminary comparison.

- With regard to the systems in Figs. 3 and 4, auxiliary consumptions have been calculated based on reasonable pressure heads across the blowers (6-10 kPa) and on isentropic and mechanical-electrical efficiency values of 0.75 and 0.9, respectively.
- The energy demand related to the oxygen production by PSA/VSA technology in Fig. 4 has been calculated according to an electric consumption of 0.4 kWh per Nm^3 of oxygen, along with an electric conversion efficiency of 50%;
- As for the system in Fig. 5, pressure ratios of 4.43 and 1.1 for the vacuum pump C1 and the blower C2 have been considered, along with isentropic and mechanical-electrical efficiency values of 0.75 and 0.9, respectively; pressure ratios of 4.2 and 3.88 with isentropic efficiency values of 0.83 and 0.87 have been considered for the compressor C3 and the turbine T1, respectively, with the mechanical-electrical efficiency fixed at 0.9.

Table 2. Details of fuel and electricity consumptions for the examined cases (1: reference case; 2: oxy-fuel case with heat recovery; 3: oxy-fuel case with oxygen transport membrane).

	1	2	3
Fuel to the glass melting furnace, kg/h (kW)	309.3 (3992.7)	235.7 (3043.5)	234.4 (3026.6)
Electric power duty due to auxiliaries, kW	105.1	47.3	77.1
Electric power duty for O_2 production, kW	-	275.2	-
Electric power from the micro-gas turbine, kW	-	-	-319.6
Overall electric power, kW	105.1	322.5	-242.5
Equivalent fuel, kg/h	16.3	50.0	-37.6
Additional fuel, kg/h	-	-	57.1
Primary fuel, kg/h (kW)	325.6 (4202.9)	285.7 (3688.5)	254.0 (3280)

A quick overview of the energy consumption results is reported in Table 2. The net power from the micro-gas turbine is negative as it is a system output. Equivalent fuel values are reported based on the overall (positive or negative) electric power and an electric conversion efficiency of 50%. Thus, an assessment of the primary fuel energy demand is possible for all three cases considered in this paper. Paying attention to the primary fuel results, it is possible to realize the superiority of the oxy-fuel solutions, from an energy saving point of view, especially in the case of the advanced membrane-based technology.

4. Economic considerations

This section deals with considerations about the possible economic benefits in oxy-fuel glass melting furnaces with oxygen production via a membrane-based technology instead of conventional solutions. Referring to the results in Table 2, which details the energetic performance of the investigated systems, a comparison between the two oxy-fuel cases is here proposed based on relatively basic calculations. In particular, a differential comparison is made since the glass melting furnace is always the same, despite the different oxygen production solutions.

As regards the conventional oxygen production technology (based on pressure or vacuum swing adsorption), investment costs (C) of 5 and 1 M€ for plant capacity (Q) of 2000 Nm^3/h and one order of magnitude lower (~200 Nm^3/h), respectively, are considered in agreement to the authors' knowledge. Assuming an exponential trend in investment cost according to capacity ratio as $C = C_0 \cdot (Q/Q_0)^f$, an exponential factor f = 0.7 can be inferred from the previous values. Considering the investigated case in Fig. 4 requires around 630 Nm^3/h of pure oxygen, it is possible to estimate 2228.6 k€ as the total cost of the plant for oxygen production via conventional technology.

Referring to the advanced case in Fig. 5, component costs have to be better broken down. As a matter of fact, there are metallic recuperators as well as one turbine and some blowers/compressors with a strong system integration. Reference to the methodology proposed by Galanti and Massardo [8] is made here for a quick estimation of these component costs, so the direct material costs for the micro-gas turbine system amounts to 491 k€ (a cost of 210 k€ for a 200 kW micro-gas turbine has been considered). This cost is increased by 30% to take customization into account and further 65.6 k€ are considered for additional recuperative heat exchangers and the oxygen vacuum pumping equipment. The sum of such direct material costs is then increased to include EPC services (32%), construction costs (20%) and other costs (8%), resulting in 1126.2 k€.

In order to include the membrane system, a specific cost of 1900 €/m^2 is assumed [9]. Based on the calculated membrane area (402.1 m^2), the cost for the membranes results 763.9 k€, plus 10% as the estimated cost for the reactor. The resulting direct material costs for the membrane system will then be increased to include EPC services (16%), construction (10%) and other costs (8%). Thus, the total costs for the membrane system come to 1126 k€, which ultimately result in 2252.2 k€ after including the previously calculated costs for the micro-gas turbine system and the additional recuperative heat exchangers.

On the other hand, with reference to the case in Fig. 4 and always according to the adopted methodology [8], further 93.8 k€ should be considered for the cost of the components present in the layout of Fig. 5 (but not included in the one of Fig. 5), i.e. recuperative heat exchangers and blowers. Such direct material costs ultimately come to 150.1 k€ as the total costs for additional plant engineering.

The total plant costs of the two compared technologies, excepting the glass melting furnace and the after-treatment station, do not seem to differ significantly at the moment.

In order to propose a preliminary economic assessment, the following assumptions have been made: 10-year plant life, 8% discount rate, energy valued at the usual prices of the Italian market (0.79 €/Nm^3 for natural gas, 0.159 €/kWh for the electricity), as well as 8000 h of plant operation per year. No maintenance costs are considered for the sake of simplicity, even though the membranes could be subject to fouling as well as occlusion.

Table 3. Cost assessment results for the two considered oxy-fuel cases

	conventional O_2 production	advanced O_2 production
Cost of plant*, k€	2378.7	2252.2
Yearly cost of fuel for the furnace, k€	1849.4	1839.4
Yearly cost of additional fuel, k€	-	448.3
Yearly cost of electricity, k€	320.8	-406.6
Discounted total cost, M€	16.94	14.86

*excepting the glass melting furnace and the after-treatment station

Considering that the net present value of 10 unit payments discounted with the assumed rate is equal to 6.71, cost calculation results for the two cases are reported in Table 3, which further remark the energy superiority previously highlighted in Table 2 for the case with the proposed advanced oxygen production.

5. Conclusions

An original work has been proposed for the possible application of oxygen transport membranes in glass melting furnaces.

Starting from a benchmark, air-blown case, the mass and energy balances have been reproduced also for two oxy-fuel glass furnaces: the first with oxygen produced by conventional technology and the second where the oxygen is separated from a hot air stream in a membrane module integrated with a micro-gas turbine.

The assessment of the primary fuel demand of the furnaces highlights the superiority of the oxy-fuel technology compared to the air-blown reference case. In detail, the solution here proposed with the advanced membrane-based technology is very promising from an energy saving point of view. Economic considerations further strengthen this conclusion.

Acknowledgements

This work was supported by the Research Fund for the Electrical System in the context of the Program Agreement between RSE S.p.A. and the Ministry of Economic Development - Management for nuclear, renewable energies and energy efficiency - in compliance with the Ministerial Decree of March 8, 2006.

References

[1] Sardeshpande V., Gaitonde U.N., Banerjee R., Model based energy benchmarking for glass furnace, Energy Conversion and Management 2007;48:2718-2738

[2] Beerkens R.G.C., van Limpt J., Energy efficiency benchmarking of glass furnaces. In: Proceedings of 62nd Conference on glass problems, University of Illinois, Urbana-Campaign, Illinois, October 16-17, 2001, DOI: 10.1002/9780470294727.ch7

[3] Beerkens R., Concepts for energy & emission friendly glass melting: Evolution or revolution in glass melting, In: Proceedings of 9th International Conference on Advances in Fusion and Processing of Glass, Cairns, Australia, July 11-14, 2011. Available at: <http://publications.tno.nl/publication/100347/EAHeqQ/beerkens-2012-new.pdf> [accessed 19.5.2018].

[4] GS (Gas-Steam cycles) – Available at: <http://www.gecos.polimi.it/software/gs.php> [accessed 19.5.2018].

[5] Mastropasqua L., Clean energy conversion systems - High temperature solid oxide electrochemical membranes in advanced power generation applications, Ph.D. Thesis, Politecnico di Milano, Italy, 2017

[6] Beerkens R.G.C., Muysenberg H.P.H., Comparative study on energy-saving technologies for glass furnaces, Glastechnische Berichte 1992;65(8):216-224

[7] An innovative oxygen combustion technology for environment friendly primary flat glass production – Available at:<http://www.oxyfuel-heatrecovery.com> [accessed 19.5.2018]

[8] Galanti L., Massardo A.F., Micro gas turbine thermodynamic and economic analysis up to 500 kWe size, Applied Energy 2011;88:4795-4802

[9] DEMOYS Publishable Summary – Available at:<https://cordis.europa.eu/docs/results/241/241309/final1-demoys-publishable-summary-report-re1.pdf> [accessed 19.5.2018]